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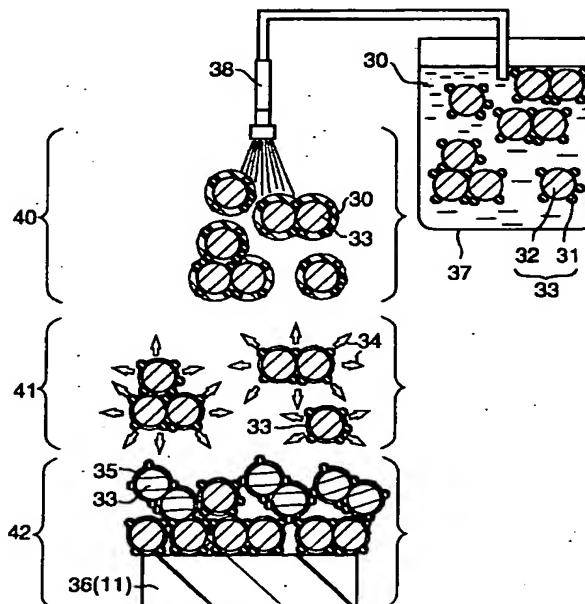
(54)【発明の名称】 燃料電池の電極触媒層形成方法

(57)【要約】

【課題】 プロトン伝導性とガス拡散性がバランスよく成立する電極触媒層を形成できる燃料電池電極触媒層形成方法の提供。

【解決手段】 (1)触媒担持粒子33混合電解質溶液30を空中にスプレーし、空中で触媒担持粒子周囲の電解質溶液の溶媒を一部揮発させ、半固体状態の電解質にて覆われた触媒担持粒子33を被塗着物36に塗着させる燃料電池の電極触媒層形成方法であって、触媒担持粒子混合電解質溶液30を複数回スプレーして各スプレーによる層を被塗着物36上に塗り重ね、各スプレー毎に層を乾燥させ、スプレー毎に液体組成、触媒、たとえば、電解質35の量を異ならせた方法。(2)被塗着物36に近い層側が電解質溶液中の電解質35の量が多い。

(3)被塗着物36を燃料電池の電解質膜11とする。
(4)スプレーにまたはスプレー周りに加温された溶液拡散用エアを流して乾燥を促進させる。



【特許請求の範囲】

【請求項1】触媒担持粒子が混合された電解質溶液を空中にスプレーし、空中で前記触媒担持粒子周囲の電解質溶液の溶媒を一部揮発させ、半固体状態の電解質にて覆われた触媒担持粒子を被塗着物に塗着させる燃料電池の電極触媒層形成方法であって、前記触媒担持粒子が混合された電解質溶液を複数回スプレーして各スプレーによる層を前記被塗着物上に塗り重ね、各スプレー毎に層を乾燥させ、かつ各スプレーでスプレーする液体組成、触媒を異ならせた燃料電池の電極触媒層形成方法。

【請求項2】触媒担持粒子が混合された電解質溶液を空中にスプレーし、空中で前記触媒担持粒子周囲の電解質溶液の溶媒を一部揮発させ、半固体状態の電解質にて覆われた触媒担持粒子を被塗着物に塗着させる燃料電池の電極触媒層形成方法であって、前記触媒担持粒子が混合された電解質溶液を複数回スプレーして各スプレーによる層を前記被塗着物上に塗り重ね、各スプレー毎に層を乾燥させ、かつ各スプレーで電解質の量を異ならせた請求項1記載の燃料電池の電極触媒層形成方法。

【請求項3】前記被塗着物に近い層側が電解質溶液中の電解質の量が多い請求項2記載の燃料電池の電極触媒層形成方法。

【請求項4】前記被塗着物を燃料電池の電解質膜とし該電解質膜上に直接電極触媒層を形成する請求項1または請求項2または請求項3記載の燃料電池の電極触媒層形成方法。

【請求項5】触媒担持粒子が混合された電解質溶液を空中にスプレーし、空中で前記触媒担持粒子周囲の電解質溶液の溶媒を一部揮発させ、半固体状態の電解質にて覆われた触媒担持粒子を被塗着物に塗着させる燃料電池の電極触媒層形成方法であって、前記被塗着物を燃料電池の電解質膜とし電解質膜上に直接電極触媒層を形成する燃料電池の電極触媒層形成方法。

【請求項6】触媒担持粒子が混合された電解質溶液を空中にスプレーするとともにスプレーされた触媒担持粒子混合電解質溶液にまたはスプレーされた触媒担持粒子混合電解質溶液周りに溶液拡散用ガスを流し、空中で前記触媒担持粒子周囲の電解質溶液の溶媒を一部揮発させ、半固体状態の電解質にて覆われた触媒担持粒子を被塗着物に塗着させる燃料電池の電極触媒層形成方法であって、前記溶液拡散用ガスを加温されたガスとした燃料電池の電極触媒層形成方法。

【発明の詳細な説明】

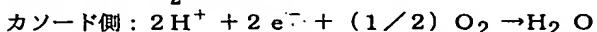
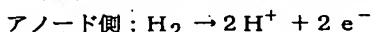
【0001】

【発明の属する技術分野】本発明は、燃料電池、とくに固体高分子電解質型燃料電池の、電極触媒層形成方法に関する。

【0002】

【従来の技術】固体高分子電解質型燃料電池は、イオン交換膜からなる電解質膜（基本的には電気絶縁体）とこ

の電解質膜の一面に配置された触媒層および拡散層からなる電極（アノード、燃料極）および電解質膜の他面に配置された触媒層および拡散層からなる電極（カソード、空気極）とからなる膜一電極アッセンブリ（MEA : Membrane-Electrode Assembly）と、アノード、カソードに燃料ガス（水素）および酸化ガス（酸素、通常は空気）を供給するための流体通路を形成するセパレータとからセルを構成し、複数のセルを積層してモジュールとし、モジュールを積層してモジュール群を構成し、モジュール群のセル積層方向両端に、ターミナル、インシユレータ、エンドプレートを配置してスタックを構成し、スタックをセル積層体積層方向に締め付け、セル積層体積層方向に延びる締結部材（たとえば、テンションプレート）にて固定したものからなる。固体高分子電解質型燃料電池では、アノード側では、水素を水素イオンと電子にする反応が行われ、水素イオンは電解質膜中をカソード側に移動し、カソード側では酸素と水素イオンおよび電子（隣りのMEAのアノードで生成した電子がセパレータを通してくる、または外部電気的負荷を通してくる）から水を生成する反応が行われる。



燃料電池電極の触媒層には、電気伝導性、プロトン伝導性、ガス拡散性がバランスよく成立する構造が求められる。従来の電極触媒層の形成方法は、特開平8-88008号公報に開示されているように、あるいは図7に示すように、湿式コーティングが一般的であり、かつ電解質膜に直接形成すると電解質膜に収縮が発生するため、転写基材（ポリテトラフルオロエチレンシート）4に触媒層5を塗布し（図7の工程1）ついで乾燥し（図7の工程2）、それを電解質膜6に熱圧着し、転写基材を剥がすことにより、電解質膜6に触媒層5を転写している（図7の工程3）。とくに特開平8-88008号公報は、燃料電池の電極触媒層で膜側の電解質質量が電極側の電解質質量より大としたものを開示している。

【0003】

【発明が解決しようとする課題】しかし、上記湿式コーティング法には、プロトン伝導性とガス拡散性がバランスよく成立する電極触媒層構造を作成することが難しいという問題がある。その理由は、以下の通りである。触媒、電解質、溶媒の混合懸濁液をコーティングした場合、塗布直後は図7の工程1に示すように、電解質は溶液状態のままである。塗布後乾燥状態では、図7の工程2に示すように、触媒を覆う電解質の厚さが均等でなく、プロトン伝導に無駄な厚さ大の電解質部分が多く、この無駄な電解質部分がガス流路を塞ぎガス拡散性を悪化させている。また、電解質が沈降し、下部（ポリテトラフルオロエチレンシート側）の電解質密度が濃くなっている。転写状態では、図7の工程3に示すように、ガスの入口である反電解質膜側が電解質で塞がれガス拡散

性が低下して性能低下を招き、プロトンの入口である電解質膜側の電解質量が少なく、電解質膜からのプロトン伝導性が悪くなる。プロトン伝導性を優先して電解質量を多くすると、無駄な電解質部分が多くなりそれがガス流路を塞ぎ、ガス拡散性が悪くなる。逆に、ガス拡散性を優先して電解質量を少なくすると、電解質膜に転写した時に電解質膜に接する側の電解質が少くなり、プロトン伝導性が悪くなり、燃料電池の性能が低下する。したがって、プロトン伝導性とガス拡散性がバランスよく成立する電極触媒層構造を形成することは困難であった。また、特開平8-88008号公報の製造方法では、磁力や遠心力で触媒担持カーボンを偏らせるため、電解質が偏在するおそれがあった。本発明の目的は、プロトン伝導性とガス拡散性がバランスよく成立する電極触媒層を形成できる燃料電池電極触媒層形成方法を提供することにある。

【0004】

【課題を解決するための手段】上記目的を達成する本発明はつぎの通りである。

(1) 触媒担持粒子が混合された電解質溶液を空中にスプレーし、空中で前記触媒担持粒子周囲の電解質溶液の溶媒を一部揮発させ、半固体状態の電解質にて覆われた触媒担持粒子を被塗着物に塗着させる燃料電池の電極触媒層形成方法であって、前記触媒担持粒子が混合された電解質溶液を複数回スプレーして各スプレーによる層を前記被塗着物上に塗り重ね、各スプレー毎に層を乾燥させ、かつ各スプレーでスプレーする液体組成、触媒を異ならせた燃料電池の電極触媒層形成方法。

(2) 触媒担持粒子が混合された電解質溶液を空中にスプレーし、空中で前記触媒担持粒子周囲の電解質溶液の溶媒を一部揮発させ、半固体状態の電解質にて覆われた触媒担持粒子を被塗着物に塗着させる燃料電池の電極触媒層形成方法であって、前記触媒担持粒子が混合された電解質溶液を複数回スプレーして各スプレーによる層を前記被塗着物上に塗り重ね、各スプレー毎に層を乾燥させ、かつ各スプレーで電解質の量を異ならせた(1)記載の燃料電池の電極触媒層形成方法。

(3) 前記被塗着物に近い層側が電解質溶液中の電解質の量が多い(2)記載の燃料電池の電極触媒層形成方法。

(4) 前記被塗着物を燃料電池の電解質膜とし該電解質膜上に直接電極触媒層を形成する(1)または(2)または(3)記載の燃料電池の電極触媒層形成方法。

(5) 触媒担持粒子が混合された電解質溶液を空中にスプレーし、空中で前記触媒担持粒子周囲の電解質溶液の溶媒を一部揮発させ、半固体状態の電解質にて覆われた触媒担持粒子を被塗着物に塗着させる燃料電池の電極触媒層形成方法であって、前記被塗着物を燃料電池の電解質膜とし電解質膜上に直接電極触媒層を形成する燃料電池の電極触媒層形成方法。

(6) 触媒担持粒子が混合された電解質溶液を空中にスプレーするとともにスプレーされた触媒担持粒子混合電解質溶液にまたはスプレーされた触媒担持粒子混合電解質溶液周りに溶液拡散用ガスを流し、空中で前記触媒担持粒子周囲の電解質溶液の溶媒を一部揮発させ、半固体状態の電解質にて覆われた触媒担持粒子を被塗着物に塗着させる燃料電池の電極触媒層形成方法であって、前記溶液拡散用ガスを加温されたガスとした燃料電池の電極触媒層形成方法。

- 10 【0005】上記(1)の燃料電池の電極触媒層形成方法では、各スプレー毎に層を乾燥させ、かつ各スプレーでスプレーする液体組成、触媒を異ならせたので、層毎に組成が異なる電極触媒層を確実に形成でき、プロトン伝導性とガス拡散性がバランスよく成立する電極触媒層の形成が可能になる。上記(2)の燃料電池の電極触媒層形成方法では、各スプレー毎に層を乾燥させ、かつ各スプレーで電解質の量を異ならせたので、層毎に電解質量が異なる電極触媒層を確実に形成でき、プロトン伝導性とガス拡散性がバランスよく成立する電極触媒層の形成が可能になる。上記(3)の燃料電池の電極触媒層形成方法では、被塗着物に近い層側程、電解質溶液中の電解質の量を多くしたので、被塗着物が電解質膜である場合、電解質膜に近い側程、電解質の量が多い触媒層を形成でき、プロトン伝導性とガス拡散性がバランスよく成立する。上記(4)または(5)の燃料電池の電極触媒層形成方法では、電解質膜上に直接電極触媒層を形成するので、従来のようにポリテトラフルオロエチレンシートに触媒層を形成しそれを電解質膜に転写する必要がなくなる。上記(6)の燃料電池の電極触媒層形成方法では、スプレーされた触媒担持粒子混合電解質溶液に、またはスプレーされた触媒担持粒子混合電解質溶液周りに溶液拡散用ガスを流し、該溶液拡散用ガスを加温されたガスとしたので、加温されたガスにより、スプレーされた触媒担持粒子混合電解質溶液中の溶媒成分の気化が促進されて、被塗着物に塗着された後での乾燥が少なくて、被塗着物の収縮、しわの発生が抑制される。また、被塗着物に塗着された後での気化が少ないので、塗布部周辺の発火防止にもなる。

【0006】

- 40 【発明の実施の形態】以下に、本発明実施例の燃料電池の電極触媒層形成方法を、図1～図6を参照して、説明する。本発明実施例の燃料電池の電極触媒層形成方法が適用される燃料電池は、固体高分子電解質型燃料電池10である。この燃料電池10は、たとえば燃料電池自動車に搭載される。ただし、自動車以外に用いられてもよい。

【0007】固体高分子電解質型燃料電池10は、図5、図6に示すように、イオン交換膜からなる電解質膜11(基本的には、電気絶縁体)とこの電解質膜11の一面に配置された触媒層12および拡散層13からなる

電極14(アノード、燃料極)および電解質膜11の他面に配置された触媒層15および拡散層16からなる電極17(カソード、空気極)とからなる膜-電極アセンブリ(MEA:Membrane-Electrode Assembly)と、電極14、17に燃料ガス(水素)および酸化ガス(酸素、通常は空気)を供給するための反応ガス流路27

(単に、ガス流路ともいう)および燃料電池冷却用の冷媒(通常は冷却水)が流れる冷媒流路26(冷却水流路ともいう)を形成するセパレータ18とからセルを形成し、少なくとも1層のセルからモジュール19を形成し、モジュール19を積層してモジュール群を構成し、モジュール19群のセル積層方向両端に、ターミナル20、インシュレータ21、エンドプレート22を配置してセル積層体を構成し、セル積層体をセル積層方向に締め付け、エンドプレート22をセル積層体の外側でセル積層体積層方向に延びる締結部材24(たとえば、テンションプレート)とボルト25で固定して、スタック23としたものからなる。

【0008】本発明実施例の燃料電池の電極触媒層形成方法は、図1、図2に示すように、固体の触媒31を担持した固体の粒子32(粒子は複数の粒子の集合からなる粒子群である場合を含む)からなる触媒担持粒子33が混合、懸濁された電解質溶液30(電解質35を溶媒で溶かした溶液)を空中にスプレーする工程40と、空中で触媒担持粒子33周囲の電解質溶液30の溶媒を一部揮発させる工程41と(34が揮発分)、揮発によって一部の溶媒が抜けることによって半固体状態となった電解質35にて覆われた触媒担持粒子33を被塗着物36に塗着させる工程42と、からなる。被塗着物36は、望ましくは固体高分子電解質型燃料電池10の電解質膜11であるが、固体高分子電解質型燃料電池10の電極拡散層であってもよい。被塗着物36が固体高分子電解質型燃料電池10の電解質膜11である場合は、従来のように電極触媒層をポリテトラフルオロエチレンシート上に形成しそれを電解質膜に転写するのではなく、電極触媒層が電解質膜11上に直接形成されることになる。

【0009】上記電極触媒層形成方法では、触媒31はたとえばPt(白金)であり、粒子32はたとえばカーボン粒子であり、電解質35および電解質膜11はたとえばフッ素系スルホン酸高分子樹脂で、一例としてナフイオン(デュポン社製の商品名)がある。「スプレー」は、「噴霧」であってもよいし「散布」であってもよく、スプレーされたものが、空中で霧状または粒状になればよい。スプレーは容器37内の電解質溶液をポンプ等にてノズル38からスプレーすることにより行う。

【0010】この電極触媒層形成方法では、スプレー工程40で、空中の触媒担持粒子33は、周囲の電解質溶液30の表面張力により周囲の電解質溶液30で均一に覆われた状態になる。この状態でさらに空中を飛ばすと

(工程41)、触媒担持粒子33周囲の電解質溶液30の溶媒が一部蒸発し、均一に覆った状態を維持しつつ、半固体状態となる。この状態で電解質膜11上に塗着されると、触媒担持粒子33周囲に電解質35が均一に覆った状態で積層し、かつ無駄な電解質が少ないので、触媒層中に多孔が形成される(工程42)。触媒担持粒子33を覆う電解質33の量、厚さは、電解質溶液30中の電解質量を増減させることにより変化させ制御することができる。上記スプレー法による電極触媒層形成方法10によって、触媒担持粒子33を周囲の電解質溶液30で均一に覆った電極触媒層が得られ、したがって、理想的な3相界面が得られ、プロトン伝導性とガス拡散性を高いレベルで両立させることができ、なおかつ、その電解質量を制御できる。

【0011】電解質溶液30中の溶媒は、低沸点のものを使用することが望ましい。そうすることによって、空中での溶媒の揮発速度が速いため、ノズル38と電解質膜11の距離を小さ目にすることができ、周囲に飛散して無駄になる触媒量を減らすことができる。

【0012】触媒担持粒子33が混合、懸濁された電解質溶液30を複数回スプレーして各スプレーによる層を被塗着物36(電解質膜11)上に塗り重ねて多層塗りとしてもよい。その場合、各スプレー毎に層を乾燥させ、かつ各スプレーで電解質溶液30中の電解質35の量を異ならせる。この多層塗りによって、1層の塗布量を少なくすることができ、塗着後の電解質溶液の乾燥が促進され、空中での溶媒の揮発が不十分な場合でも、塗着後の電解質皮膜の変形を最小限とすることができる。

【0013】この複数回スプレーによる多層塗りにおいて、1回毎のスプレーの電解質溶液30中の液体組成、触媒を変化させることができ、たとえば、電極層厚さ方向に電解質量を変化させることができる。たとえば、図3に示すように、移動する電解質膜11上に複数の容器37を配置し、容器37毎に電解質溶液30中の電解質量を変える。この場合、電解質膜11側の電解質量をリッチに、反電解質膜側の電解質量をプアにする。これによつて、プロトンは電解質膜から電極に移動し徐々に消費されていき、また反応ガスは反電解質膜側から徐々に消費されていくため、さらに効率の高い電極構造が得られる。

【0014】上記スプレー法による電極形成は、電解質膜11上への、直接の電極触媒層の形成に適用されてもよい。そうすることによって、ポリテトラフルオロエチレンシートにスプレー法によって触媒層を形成しそれを電解質膜に転写する場合に比べて、工程減をはかることができる。その場合は、ポリテトラフルオロエチレンシートにスプレー法によって触媒層を形成しそれを電解質膜に転写する場合に生じる、スプレー形成電極の表面の比較的大きな凹凸による、スタック締め付け後の電解質膜のクリープと、それによるアノード、カソード間の電

解質膜における電気的微量短絡、の発生のおそれを除去することができる。上記は電解質膜11上に電極層を形成する場合であったが、カーボン多孔生地に本発明のスプレー法によって電極拡散層や、電極拡散層と触媒層を形成してもよい。

【0015】また、図4に示すように、上記スプレー法において、触媒担持粒子33が混合された電解質溶液30を空中にスプレーするとともにスプレーされた触媒担持粒子混合電解質溶液30にまたはスプレーされた触媒担持粒子混合電解質溶液30周りに加温された溶液拡散用ガスを流し、空中で触媒担持粒子周囲の電解質溶液の溶媒を一部揮発させ、半固体状態の電解質にて覆われた触媒担持粒子を被塗着物36（たとえば、電解質膜11）に塗着させるようにしてもよい。溶液拡散用ガスは、溶液拡散用ガスノズル39から流出される。溶液拡散用ガスは、たとえばスワール生成用エアであり、スプレーされた触媒担持粒子混合電解質溶液30周りに捩じり方向にエアを流出させることにより、スプレーされた触媒担持粒子混合電解質溶液30の噴射パターンを拡げることができ、触媒担持粒子混合電解質溶液30を拡散することができる。加温された溶液拡散用ガスの温度は、被塗着物36を損傷させない程度の温度で、かつ、電解質溶液の溶媒の一部揮発に効果を発揮できる温度であり、たとえば、80℃～100℃程度である。

【0016】ノズル38に溶液拡散用エア流出ノズル43を付設し、溶液拡散用エア流出ノズル39から溶液拡散用ガス（たとえば、スワールエア）を流出させ、溶液拡散用ガスを加温しておくことにより、ノズル38先端より吐出された触媒担持粒子混合電解質溶液30の、被塗着物36に塗着する直前までの、空中における溶媒の乾燥が促進され（ただし、乾燥完了まではいかない）、被塗着物36（たとえば、電解質膜11）に塗着した後に乾燥する場合に生じる被塗着物36（たとえば、電解質膜11）の収縮、しわの発生を防止することができる。また、塗布部周辺の発火防止ともなる。

【0017】

【発明の効果】請求項1の燃料電池の電極触媒層形成方法によれば、各スプレー毎に層を乾燥させ、かつ各スプレーでスプレーする液体組成、触媒を異ならせたので、層毎に組成が異なる電極触媒層を確実に形成でき、プロトン伝導性とガス拡散性がバランスよく成立する電極触媒層を形成することができる。請求項2の燃料電池の電極触媒層形成方法によれば、各スプレー毎に層を乾燥させ、かつ各スプレーで電解質の量を異なせたので、層毎に電解質量が異なる電極触媒層を確実に形成でき、プロトン伝導性とガス拡散性がバランスよく成立する電極触媒層を形成することができる。請求項3の燃料電池の電極触媒層形成方法によれば、被塗着物に近い層側程、電解質溶液中の電解質の量を多くしたので、被塗着物が電解質膜である場合、電解質膜に近い側程、電解質の量

が多い触媒層を形成でき、プロトン伝導性とガス拡散性がバランスよく成立させることができる。請求項4の燃料電池の電極触媒層形成方法によれば、電解質膜上に直接電極触媒層を形成するので、従来のようにポリテトラフルオロエチレンシートに触媒層を形成しそれを電解質膜に転写する必要がなくなり、工程減をはかることができる。請求項5の燃料電池の電極触媒層形成方法によれば、電解質膜上に直接電極触媒層を形成するので、従来のようにポリテトラフルオロエチレンシートに触媒層を形成しそれを電解質膜に転写する必要がなくなり、工程減をはかることができる。また、カーボン生地上にスプレー法によって拡散層、触媒層を形成しそれを別途作製の電解質膜に重ねてセルを形成する場合に比べて、スタッカ締め付け後の電解質膜のクリープ、電気的微量短絡を抑制することができる。請求項6の燃料電池の電極触媒層形成方法によれば、スプレーにまたはスプレー周りに溶液拡散用ガスを流し、該溶液拡散用ガスを加温されたガスとしたので、加温されたガスにより、霧化された触媒担持粒子混合電解質溶液中の溶媒成分の気化が促進されて、被塗着物に塗着された後での乾燥が少なくなつて、被塗着物の収縮、しわの発生が抑制される。また、被塗着物に塗着された後での気化が少ないので、塗布部周辺の発火防止にもなる。

【図面の簡単な説明】

【図1】本発明実施例の燃料電池の電極触媒層形成方法における、スプレーされた触媒担持粒子およびその周囲の電解質溶液の状態を拡大して示した断面図である。

【図2】本発明実施例の燃料電池の電極触媒層形成方法における、周囲が電解質で覆われた触媒担持粒子が積層された電極触媒層の拡大断面図である。

【図3】本発明実施例の燃料電池の電極触媒層形成方法における、複数回スプレーによる多層塗りを実施している状態を示した斜視図である。

【図4】本発明実施例の燃料電池の電極触媒層形成方法における、スプレー周りに溶液拡散用ガスを流している状態を示した斜視図である。

【図5】本発明実施例の燃料電池の電極触媒層形成方法が適用される燃料電池の正面図である。

【図6】本発明実施例の燃料電池の電極触媒層形成方法が適用される燃料電池の、一部拡大断面図である。

【図7】従来の燃料電池の電極触媒層形成方法における、コーティングされた触媒担持粒子およびその周囲の電解質溶液の状態を、工程順に拡大して示した断面図である。

【符号の説明】

10 (固体高分子電解質型) 燃料電池

11 電解質膜

12 触媒層

13 拡散層

14 電極 (アノード、燃料極)

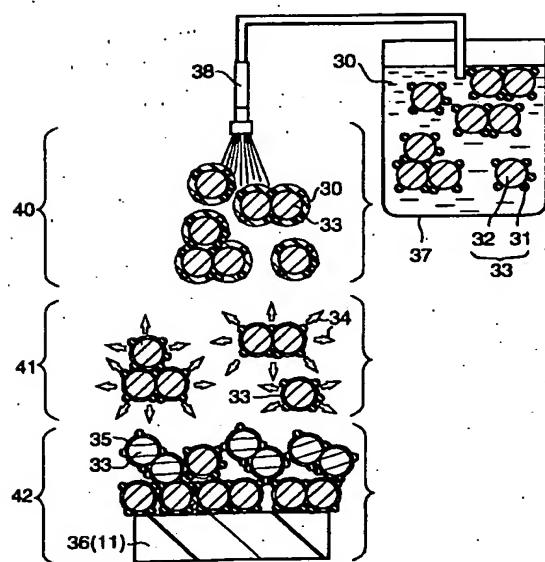
9

- 1 5 触媒層
 1 6 拡散層
 1 7 電極(カソード、空気極)
 1 8 セパレータ
 1 9 モジュール
 2 0 ターミナル
 2 1 インシュレータ
 2 2 エンドプレート
 2 3 スタック
 2 4 テンショシプレート
 2 5 ボルト
 2 6 冷媒流路

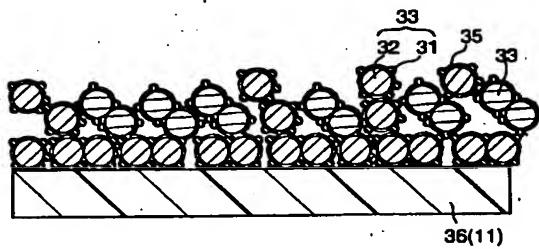
10

- 2 7 ガス流路
 3 0 電解質溶液
 3 1 触媒
 3 2 粒子
 3 3 触媒担持粒子
 3 4 挥発分
 3 5 電解質
 3 6 被塗着物
 3 7 容器
 10 3 8 ノズル
 3 9 溶液拡散用ガスノズル
 4 0、4 1、4 2 工程

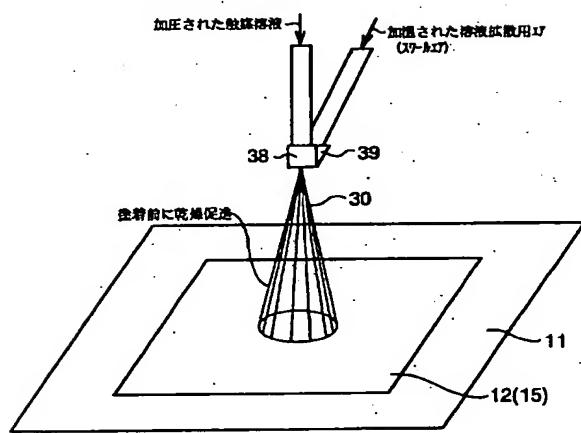
【図1】



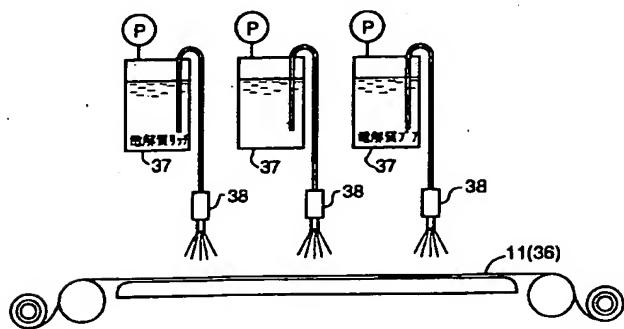
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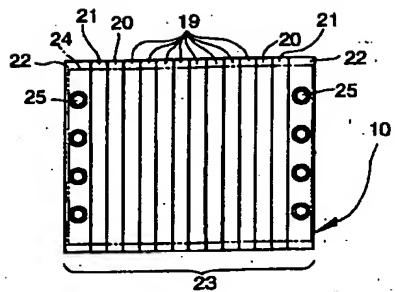
【図4】



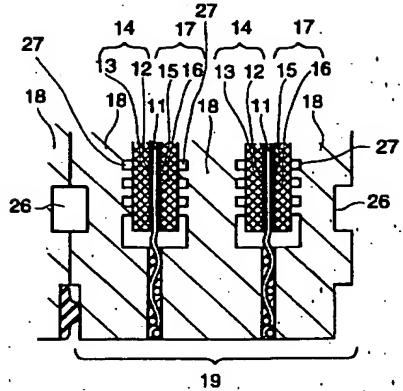
【図3】



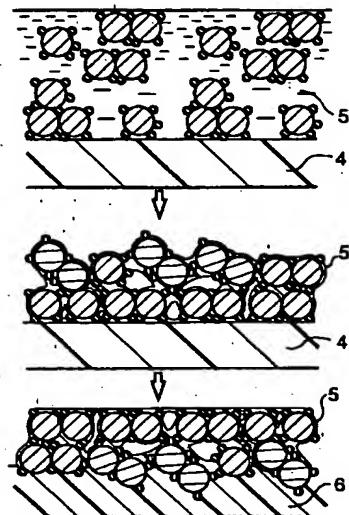
【図5】



【図6】



【図7】



フロントページの続き

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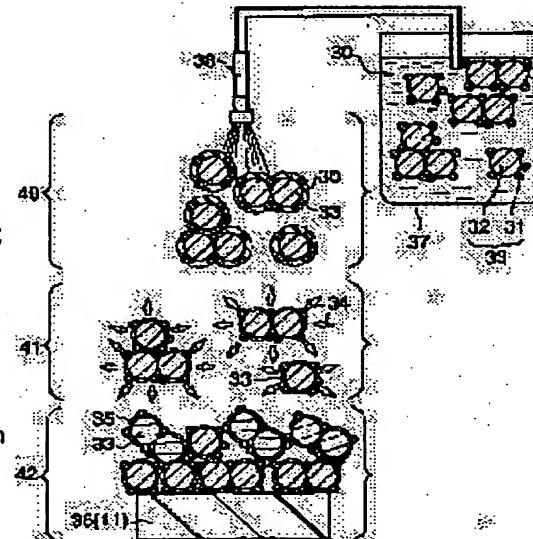
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(54) METHOD FOR FORMING ELECTRODE CATALYST LAYER OF FUEL CELL

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a method for forming a fuel cell electrode catalyst layer, capable of forming an electrode catalyst layer which is well-balanced in proton conductivity and gas diffusion.

SOLUTION: The method includes (1) catalyst carrier particle 33 mixed electrolyte solution 30 is sprayed in the air, to volatilize a part of solvent of the electrolyte solution around the catalyst carrier particle 33 in the air; the catalyst carrier particle 33, covered with semisolid electrolyte, is coated on a matter to be coated 36; catalyst carrier particle 33 mixed electrolyte solution 30 is sprayed plural number of times, and a layer of each spraying is recoated on the matter to be coated 36 and the layer is dried at each spraying; the liquid composition catalyst for example, the quantity of an electrolyte 35 is made to be different between each spraying; (2) the quantity of the electrolyte 35 in electrolyte solution is large in a layer close to the matter to be coated 36; (3) the matter to be coated 36 is used as an electrolyte film 11 of the fuel cell; and (4) heated air for diffusing solution is sent to the spray or a vicinity of the spray, to accelerate drying.



[Date of final disposal for application]

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[Date of registration]

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[Date of extinction of right]

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CLAIMS

[Claim(s)]

[Claim 1] Carry out the spray of the electrolytic solution with which a catalyst support particle was mixed in the air, and a part of solvent of an electrolytic solution of said perimeter of a catalyst support particle is volatilized in the air. It is the electrode catalyst bed formation method of a fuel cell of making a plastered object plastering with a catalyst support particle covered with an electrolyte of a semisolid condition. A liquid presentation which carries out the multiple-times spray of the electrolytic solution with which said catalyst support particle was mixed, recoats a layer by each spray to said plastered lifter, and is made to dry a layer for every spray, and carries out a spray by each spray, an electrode catalyst bed formation method of a fuel cell of having changed a catalyst.

[Claim 2] Carry out the spray of the electrolytic solution with which a catalyst support particle was mixed in the air, and a part of solvent of an electrolytic solution of said perimeter of a catalyst support particle is volatilized in the air. It is the electrode catalyst bed formation method of a fuel cell of making a plastered object plastering with a catalyst support particle covered with an electrolyte of a semisolid condition. An electrode catalyst bed formation method of a fuel cell according to claim 1 of having carried out the multiple-times spray of the electrolytic solution with which said catalyst support particle was mixed, having recoated a layer by each spray to said plastered lifter, and having dried a layer for every spray, and having changed an electrolytic amount by each spray.

[Claim 3] An electrode catalyst bed formation method of a fuel cell according to claim 2 with many [a layer side near said plastered object] amounts of an electrolyte in an electrolytic solution.

[Claim 4] An electrode catalyst bed formation method of a fuel cell according to claim 1, 2, or 3 which uses said plastered object as an electrolyte film of a fuel cell, and forms a direct electrode catalyst bed on this electrolyte film.

[Claim 5] The electrode catalyst-bed formation method of a fuel cell which a part of solvent of an electrolytic solution of said perimeter of a catalyst support particle is volatilized in the air, and is the electrode catalyst-bed formation method of a fuel cell of making a plastered object plastering with a catalyst support particle covered with an electrolyte of a semisolid condition, uses [the spray of the electrolytic solution with which a catalyst support particle was mixed is carried out in the air, and] said plastered object as an electrolyte film of a fuel cell, and forms a direct electrode catalyst bed on an electrolyte film.

[Claim 6] a catalyst support particle mixing electrolytic solution by which the spray was carried out while carrying out the spray of the electrolytic solution with which a catalyst support particle was mixed in the air -- or the circumference of a catalyst support particle mixing electrolytic solution by which the spray was carried out -- gas for solution diffusion -- a sink -- An electrode catalyst bed formation method of a fuel cell made into gas which it is [gas] the electrode catalyst bed formation method of a fuel cell of making a plastered object plastering with a catalyst support particle which a part of solvent of an electrolytic solution of said perimeter of a catalyst support particle was volatilized in the air, and was covered with an electrolyte of a semisolid condition, and had said gas for solution diffusion warmed.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] This invention relates to the electrode catalyst bed formation method of a fuel cell, especially a solid-state polyelectrolyte mold fuel cell.

[0002]

[Description of the Prior Art] the electrode (an anode --) which consists of the catalyst bed and diffusion layer which have been arranged at the whole surface of the electrolyte film (fundamentally electric insulator) with which a solid-state polyelectrolyte mold fuel cell consists of ion exchange membrane, and this electrolyte film with the film-electrode assembly (MEA:Membrane-Electrode Assembly) which consists of an electrode (a cathode, air pole) which consists of the catalyst bed and diffusion layer of a fuel electrode and an electrolyte film which were alike on the other hand and have been arranged A cel is constituted from a separator which forms the fluid channel for supplying fuel gas (hydrogen) and oxidation gas (oxygen, usually air) to an anode and a cathode. Carry out the laminating of two or more cels, consider as a module, carry out the laminating of the module, and a module group is constituted. A terminal, an insulator, and an end plate are arranged to the direction both ends of a cel laminating of a module group, a stack is constituted, a stack is bound tight in the direction of a cel layered product laminating, and it consists of what was fixed in the conclusion member (for example, tension plate) prolonged in the direction of a cel layered product laminating. With a solid-state polyelectrolyte mold fuel cell, the reaction which uses hydrogen as a hydrogen ion and an electron is performed, a hydrogen ion moves the inside of an electrolyte film to a cathode side, and the reaction which generates water from oxygen, a hydrogen ion, and an electron (the electron generated with the anode of the next MEA lets a separator pass, or lets external electrical load pass) is performed by the cathode side at an anode side.

anode side: -- $H_2 \rightarrow 2H^{++} + 2e^-$ -cathode side: -- the catalyst bed of a $2H^{++} + 2e^- + (1/2) O_2 \rightarrow H_2O$ fuel cell electrode is asked for the structure where electrical conductivity, proton conductivity, and gaseous diffusion nature are materialized with sufficient balance. Since contraction will occur on an electrolyte film if wet coating is common and forms in an electrolyte film directly, as shown in drawing 7 as the formation method of the conventional electrode catalyst bed is indicated by JP,8-88008,A or, The catalyst bed 5 is imprinted on the electrolyte film 6 by applying a catalyst bed 5 to the imprint base material (polytetrafluoroethylene sheet) 4, drying subsequently (production process 1 of drawing 7) (production process 2 of drawing 7), carrying out thermocompression bonding of it to the electrolyte film 6, and removing an imprint base material (production process 3 of drawing 7). Especially JP,8-88008,A is indicating what the amount of electrolytes by the side of a film made size from the amount of electrolytes by the side of an electrode by the electrode catalyst bed of a fuel cell.

[0003]

[Problem(s) to be Solved by the Invention] However, there is a problem that it is difficult to create the electrode catalyst bed structure where proton conductivity and gaseous diffusion nature are materialized with sufficient balance in the above-mentioned wet coating method. The reason is as follows. When the

mixed suspension of a catalyst, an electrolyte, and a solvent is coated, as shown in the production process 1 of drawing 7 immediately after spreading, an electrolyte is still a solution condition. In the dryness after spreading, as shown in the production process 2 of drawing 7, the thickness of a wrap electrolyte is not equal in a catalyst, proton conduction has many electrolyte portions of a useless thickness size, this useless electrolyte portion closes a gas passageway, and gaseous diffusion nature is worsened. Moreover, an electrolyte sediments and lower (polytetrafluoroethylene sheet side) electrolyte density is deep. In the state of an imprint, as shown in the production process 3 of drawing 7, the anti-electrolyte film side which is the entrance of gas is closed by the electrolyte, gaseous diffusion nature falls, degradation is caused, there are few amounts of electrolytes by the side of the electrolyte film which is the entrance of a proton, and the proton conductivity from an electrolyte film worsens. If priority is given to proton conductivity and the amount of electrolytes is made [many], a useless electrolyte portion will increase, it will close a gas passageway, and gaseous diffusion nature will worsen. On the contrary, if priority is given to gaseous diffusion nature and the amount of electrolytes is lessened, when it imprints on an electrolyte film, the electrolyte of the side which touches an electrolyte film will decrease, proton conductivity will worsen, and the engine performance of a fuel cell will fall. Therefore, it was difficult to form the electrode catalyst bed structure where proton conductivity and gaseous diffusion nature are materialized with sufficient balance. Moreover, by the manufacture method of JP,8-88008,A, in order to bias catalyst support carbon with magnetism or centrifugal force, there was a possibility that an electrolyte might be unevenly distributed. The purpose of this invention has proton conductivity and gaseous diffusion nature in offering the fuel cell electrode catalyst bed formation method which can form the electrode catalyst bed materialized with sufficient balance.

[0004]

[Means for Solving the Problem] This invention which attains the above-mentioned purpose is as follows.

(1) Carry out the spray of the electrolytic solution with which a catalyst support particle was mixed in the air. It is the electrode catalyst bed formation method of a fuel cell of making a plastered object plastering with a catalyst support particle which a part of solvent of an electrolytic solution of said perimeter of a catalyst support particle was volatilized in the air, and was covered with an electrolyte of a semisolid condition. A liquid presentation which carries out the multiple-times spray of the electrolytic solution with which said catalyst support particle was mixed, recoats a layer by each spray to said plastered lifter, and is made to dry a layer for every spray, and carries out a spray by each spray, an electrode catalyst bed formation method of a fuel cell of having changed a catalyst.

(2) Carry out the spray of the electrolytic solution with which a catalyst support particle was mixed in the air. It is the electrode catalyst bed formation method of a fuel cell of making a plastered object plastering with a catalyst support particle which a part of solvent of an electrolytic solution of said perimeter of a catalyst support particle was volatilized in the air, and was covered with an electrolyte of a semisolid condition. An electrode catalyst bed formation method of a fuel cell given in (1) of having carried out the multiple-times spray of the electrolytic solution with which said catalyst support particle was mixed, having recoated a layer by each spray to said plastered lifter, and having dried a layer for every spray, and having changed an electrolytic amount by each spray.

(3) An electrode catalyst bed formation method of a fuel cell given [with many / a layer side near said plastered object / amounts of an electrolyte in an electrolytic solution] in (2).

(4) (1) which uses said plastered object as an electrolyte film of a fuel cell, and forms a direct electrode catalyst bed on this electrolyte film, (2), or an electrode catalyst bed formation method of a fuel cell given in (3).

(5) The electrode catalyst-bed formation method of the fuel cell which a part of solvent of an electrolytic solution of said perimeter of a catalyst support particle is volatilized in the air, and is the electrode catalyst-bed formation method of a fuel cell of making a plastered object plastering with a catalyst support particle covered with an electrolyte of a semisolid condition, uses [the spray of the electrolytic solution with which a catalyst support particle was mixed is carried out in the air, and] said plastered object as an electrolyte film of a fuel cell, and forms a direct electrode catalyst bed on an electrolyte

film.

(6) a catalyst support particle mixing electrolytic solution by which the spray was carried out while carrying out the spray of the electrolytic solution with which a catalyst support particle was mixed in the air -- or the circumference of a catalyst support particle mixing electrolytic solution by which the spray was carried out -- gas for solution diffusion -- a sink -- An electrode catalyst bed formation method of a fuel cell made into gas which it is [gas] the electrode catalyst bed formation method of a fuel cell of making a plastered object plastering with a catalyst support particle which a part of solvent of an electrolytic solution of said perimeter of a catalyst support particle was volatilized in the air, and was covered with an electrolyte of a semisolid condition, and had said gas for solution diffusion warmed. [0005] By electrode catalyst bed formation method of a fuel cell the above (1), since a liquid presentation which is made to dry a layer for every spray, and carries out a spray by each spray, and a catalyst were changed, an electrode catalyst bed from which a presentation differs for every layer can be formed certainly, and formation of an electrode catalyst bed in which proton conductivity and gaseous diffusion nature are materialized with sufficient balance is attained. By electrode catalyst bed formation method of a fuel cell the above (2), since a layer was dried for every spray and an electrolytic amount was changed by each spray, an electrode catalyst bed from which the amount of electrolytes differs for every layer can be formed certainly, and formation of an electrode catalyst bed in which proton conductivity and gaseous diffusion nature are materialized with sufficient balance is attained. By electrode catalyst bed formation method of a fuel cell the above (3), since a layer side near a plastered object made [many] an amount of an electrolyte in an electrolytic solution, when a plastered object is an electrolyte film, a catalyst bed with more electrolytic amount to a side nearer to an electrolyte film can be formed, and proton conductivity and gaseous diffusion nature are materialized with sufficient balance. Since a direct electrode catalyst bed is formed on an electrolyte film, a catalyst bed is formed in a polytetrafluoroethylene sheet like before, and it becomes unnecessary to imprint it on an electrolyte film by the above (4) or electrode catalyst bed formation method of a fuel cell of (5). By electrode catalyst bed formation method of a fuel cell the above (6) Since gas for solution diffusion was made into gas which had a sink and this gas for solution diffusion warmed at a catalyst support particle mixing electrolytic solution by which the spray was carried out, or the circumference of a catalyst support particle mixing electrolytic solution by which the spray was carried out By warmed gas, evaporation of a solvent component in a catalyst support particle mixing electrolytic solution by which the spray was carried out is promoted, after a plastered object is plastered, desiccation decreases, and contraction of a plastered object and generating of a wrinkling are controlled. Moreover, since there is little evaporation after a plastered object is plastered, it also becomes ignition prevention of the spreading section circumference.

[0006]

[Embodiment of the Invention] Below, the electrode catalyst bed formation method of the fuel cell of this invention example is explained with reference to drawing 1 - drawing 6. The fuel cell with which the electrode catalyst bed formation method of the fuel cell of this invention example is applied is the solid-state polyelectrolyte mold fuel cell 10. This fuel cell 10 is carried in a fuel cell powered vehicle. However, it may be used in addition to an automobile.

[0007] The solid-state polyelectrolyte mold fuel cell 10 is the electrolyte film 11 (fundamentally) which consists of ion exchange membrane as shown in drawing 5 and drawing 6. the electrode 14 (an anode --) which consists of the catalyst bed 12 and diffusion layer 13 which have been arranged at the whole surface of an electric insulator and this electrolyte film 11 the electrode 17 (a cathode --) which consists of the catalyst bed 15 and diffusion layer 16 of a fuel electrode and the electrolyte film 11 which were alike on the other hand and have been arranged The film-electrode assembly which consists of an air pole (MEA:Membrane-Electrode Assembly), Reactant gas passage 27 (only) for supplying fuel gas (hydrogen) and oxidation gas (oxygen, usually air) to electrodes 14 and 17. A cel is formed from the separator 18 which forms the refrigerant passage 26 (it is also called a circulating-water-flow way) where the refrigerant for fuel cell cooling (usually cooling water) flows and it also calls it a gas passageway. Form a module 19 from the cel of at least one layer, carry out the laminating of the module

19, and a module group is constituted. To the direction both ends of a cel laminating of module 19 group, arrange a terminal 20, an insulator 21, and an end plate 22, and a cel layered product is constituted to them. A cel layered product is bound tight in the direction of a cel laminating, an end plate 22 is fixed with the conclusion member 24 (for example, tension plate) and bolt 25 which are prolonged in the direction of a cel layered product laminating on the outside of a cel layered product, and it consists of what was made into the stack 23.

[0008] The electrode catalyst bed formation method of the fuel cell of this invention example As shown in drawing 1 and drawing 2, the catalyst support particle 33 which consists of a particle 32 (a particle includes the case where it is the particle group which consists of a set of two or more particles) of the solid-state which supported the solid catalyst 31 is mixed. The production process 40 which carries out the spray of the electrolytic solution 30 (solution which melted the electrolyte 35 with the solvent) by which suspension was carried out in the air, the production process 41 which volatilizes a part of solvent of the electrolytic solution 30 of the catalyst support particle 33 perimeter in the air -- (the production process 42 which makes the plastered object 36 plaster with the catalyst support particle 33 covered with the electrolyte 35 with which 34 changed into the semisolid condition when some solvents fell out by volatile-matter) and volatilization -- since -- it becomes. Although the plastered object 36 is the electrolyte film 11 of the solid-state polyelectrolyte mold fuel cell 10 desirably, it may be the electrode diffusion layer of the solid-state polyelectrolyte mold fuel cell 10. When the plastered object 36 is the electrolyte film 11 of the solid-state polyelectrolyte mold fuel cell 10, an electrode catalyst bed is formed on a polytetrafluoroethylene sheet like before, it will not be imprinted on an electrolyte film but an electrode catalyst bed will be directly formed on the electrolyte film 11.

[0009] By the above-mentioned electrode catalyst bed formation method, a catalyst 31 is Pt (platinum), a particle 32 is for example, a carbon particle, and an electrolyte 35 and the electrolyte film 11 are for example, fluorine system sulfonic-acid macromolecule resin, and it has Nafion (Du Pont trade name) as an example. A "spray" may be the "fuel spray", may be "spraying" and should just have carried out a spray to the shape of the shape of a fog, and a grain in the air. A spray is performed by carrying out the spray of the electrolytic solution in a container 37 from a nozzle 38 with a pump etc.

[0010] In this electrode catalyst bed formation method, it will be covered with homogeneity by the catalyst support particle 33 in the air with the surrounding electrolytic solution 30 with the surface tension of the surrounding electrolytic solution 30 at the spray production process 40. If the air is further flown in this condition (production process 41), a part of solvent of the electrolytic solution 30 of the catalyst support particle 33 perimeter evaporates, and it will be in a semisolid condition, maintaining the condition of having covered to homogeneity. If it is made to plaster on the electrolyte film 11 in this condition, after the electrolyte 35 has covered to homogeneity, a laminating is carried out to the catalyst support particle 33 perimeter, and since there are few useless electrolytes, porosity will be formed into a catalyst bed (production process 42). The catalyst support particle 33 can be changed by making the amount of electrolytes in an electrolytic solution 30 fluctuate, and the amount of the wrap electrolyte 33 and thickness can control it. the electrode catalyst bed which covered the catalyst support particle 33 to homogeneity with the surrounding electrolytic solution 30 is obtained, therefore an ideal three-phase-circuit interface is acquired, and compatible [in proton conductivity and gaseous diffusion nature / on high level] by the electrode catalyst bed formation method by the above-mentioned spray method, -- it can make -- in addition -- and the amount of electrolytes is controllable.

[0011] As for the solvent in an electrolytic solution 30, it is desirable to use the thing of a low-boiling point. By doing so, since the volatilization rate of the solvent in the air is quick, distance of a nozzle 38 and the electrolyte film 11 can be made eye small **, and the amount of catalysts which disperses around and becomes useless can be reduced.

[0012] The catalyst support particle 33 carries out the multiple-times spray of mixing and the electrolytic solution 30 by which suspension was carried out, the layer by each spray is recoated on the plastered object 36 (electrolyte film 11), and it is good also as multilayer coating. In that case, a layer is dried for every spray and the amount of the electrolyte 35 in an electrolytic solution 30 is changed by each spray. By this multilayer coating, coverage of one layer can be lessened, desiccation of the

electrolytic solution after application is promoted, and even when volatilization of the solvent in the air is inadequate, deformation of the electrolyte coat after application can be made into the minimum.

[0013] In the multilayer coating by this multiple-times spray, the liquid presentation in the electrolytic solution 30 of the spray in every time and a catalyst can be changed, for example, the amount of electrolytes can be changed in the electrode layer thickness direction. For example, as shown in drawing 3, on the electrolyte film 11 which moves, two or more containers 37 are arranged and the amount of electrolytes in an electrolytic solution 30 is changed every container 37. In this case, the amount of electrolytes by the side of an anti-electrolyte film is richly made into PUA for the amount of electrolytes by the side of the electrolyte film 11. Since a proton moves to an electrode from an electrolyte film, and is gradually consumed by this and reactant gas is gradually consumed from the anti-electrolyte film side, the electrode structure where effectiveness is still higher is acquired.

[0014] The electrode formation by the above-mentioned spray method may be applied to the formation of a direct electrode catalyst bed of up to the electrolyte film 11. By doing so, the decrease of a production process can be planned compared with the case where form a catalyst bed in a polytetrafluoroethylene sheet with a spray method; and it is imprinted on an electrolyte film. In that case, fear of generating of electric minute amount short circuit ** in the electrolyte film between the creep of the electrolyte film after stack bolting by the comparatively big irregularity of the surface of a spray formation electrode produced when forming a catalyst bed in a polytetrafluoroethylene sheet with a spray method and imprinting it on an electrolyte film, the anode by it, and a cathode is removable. although the above was the case where an electrode layer was formed on the electrolyte film 11 -- carbon porosity -- an electrode diffusion layer, and an electrode diffusion layer and a catalyst bed may be formed in the ground with the spray method of this invention:

[0015] Moreover, as shown in drawing 4, it sets to the above-mentioned spray method. the catalyst support particle mixing electrolytic solution 30 by which the spray was carried out while carrying out the spray of the electrolytic solution 30 with which the catalyst support particle 33 was mixed in the air -- or the gas for solution diffusion warmed at the circumference of the catalyst support particle mixing electrolytic solution 30 by which the spray was carried out -- a sink -- You may make it make the plastered object 36 (for example, electrolyte film 11) plaster with the catalyst support particle which a part of solvent of the electrolytic solution of the perimeter of a catalyst support particle was volatilized in the air, and was covered with the electrolyte of a semisolid condition. The gas for solution diffusion flows out of the gas nozzle 39 for solution diffusion. It is air for swirl generation, and by making air flow into the circumference of the catalyst support particle mixing electrolytic solution 30 by which the spray was carried out in the torsion direction, the gas for solution diffusion can open the injection pattern of the catalyst support particle mixing electrolytic solution 30 by which the spray was carried out, and can diffuse the catalyst support particle mixing electrolytic solution 30. It is the temperature of the degree which does not damage the plastered object 36, the solvent of an electrolytic solution is the temperature which can demonstrate an effect to volatilization a part, for example, the temperature of the warmed gas for solution diffusion is 80 degrees C - about 100 degrees C.

[0016] By attaching the air outflow nozzle 43 for solution diffusion to a nozzle 38, making the gas for solution diffusion (for example, swirl air) flow out of the air outflow nozzle 39 for solution diffusion, and warming the gas for solution diffusion The catalyst support particle mixing electrolytic solution 30 breathed out from nozzle 38 tip, Desiccation of the solvent in the air until just before plastering the plastered object 36 is promoted. Contraction of the plastered object 36 (for example, electrolyte film 11) produced when drying after plastering (however, it does not go to the completion of desiccation) and the plastered object 36 (for example, electrolyte film 11), and generating of a wrinkling can be prevented. Moreover, it also becomes ignition prevention of the spreading section circumference.

[0017]

[Effect of the Invention] Since the liquid presentation which is made to dry a layer for every spray, and carries out a spray by each spray, and the catalyst were changed according to the electrode catalyst bed formation method of the fuel cell of claim 1, the electrode catalyst bed from which a presentation differs for every layer can be formed certainly, and the electrode catalyst bed in which proton conductivity and

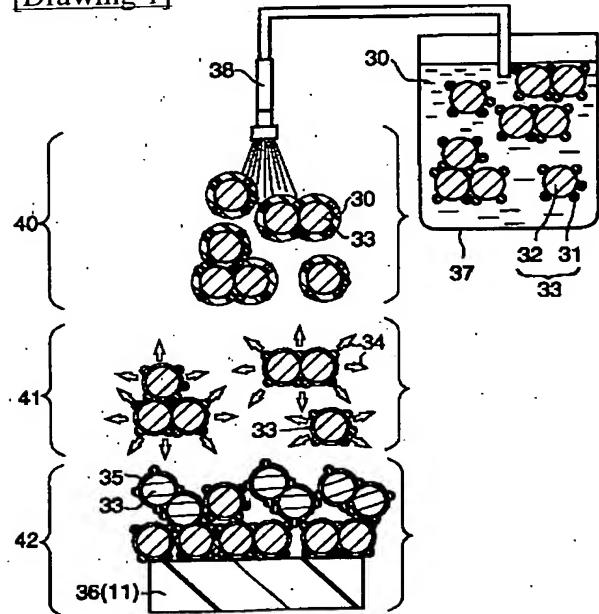
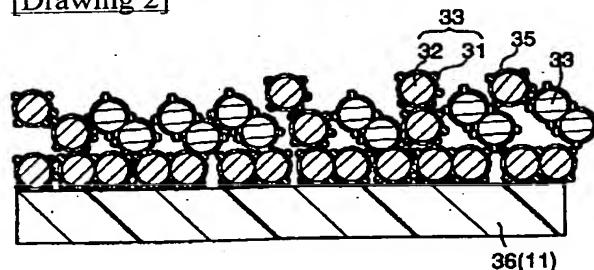
gaseous diffusion nature are materialized with sufficient balance can be formed. Since according to the electrode catalyst bed formation method of the fuel cell of claim 2 the layer was dried for every spray and the electrolytic amount was changed by each spray, the electrode catalyst bed from which the amount of electrolytes differs for every layer can be formed certainly, and the electrode catalyst bed in which proton conductivity and gaseous diffusion nature are materialized with sufficient balance can be formed. According to the electrode catalyst bed formation method of the fuel cell of claim 3, since the layer side near a plastered object made [many] the amount of the electrolyte in an electrolytic solution, when a plastered object is an electrolyte film, the side near an electrolyte film can form a catalyst bed with many electrolytic amounts, and proton conductivity and gaseous diffusion nature can form it with sufficient balance. According to the electrode catalyst bed formation method of the fuel cell of claim 4, since a direct electrode catalyst bed is formed on an electrolyte film, it becomes unnecessary to be able to form a catalyst bed in a polytetrafluoroethylene sheet like before, and to imprint it on an electrolyte film, and the decrease of a production process can be planned. According to the electrode catalyst bed formation method of the fuel cell of claim 5, since a direct electrode catalyst bed is formed on an electrolyte film, it becomes unnecessary to be able to form a catalyst bed in a polytetrafluoroethylene sheet like before, and to imprint it on an electrolyte film, and the decrease of a production process can be planned. Moreover, compared with the case where form a diffusion layer and a catalyst bed in the carbon student ground with a spray method, and a cel is formed in the electrolyte film of separately production of it in piles, the creep of the electrolyte film after stack bolting and electric fine ***** can be controlled. according to the electrode catalyst bed formation method of the fuel cell of claim 6 -- a spray -- or, since the gas for solution diffusion was made into the gas which had a sink and this gas for solution diffusion warmed at the circumference of a spray By the warmed gas, evaporation of the solvent component in the atomized catalyst support particle mixing electrolytic solution is promoted, after a plastered object is plastered, desiccation decreases, and contraction of a plastered object and generating of a wrinkling are controlled. Moreover, since there is little evaporation after a plastered object is plastered, it also becomes ignition prevention of the spreading section circumference.

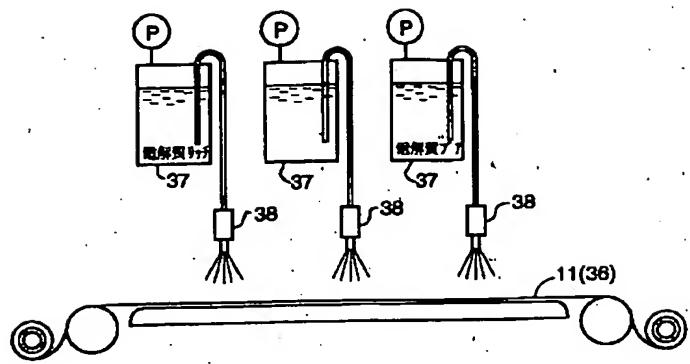
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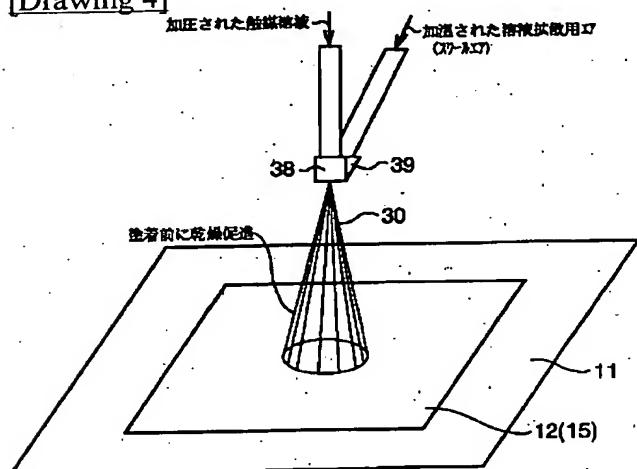
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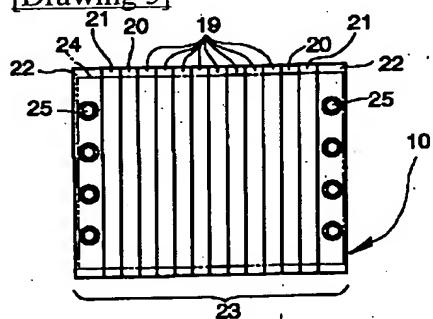
DRAWINGS**[Drawing 1]****[Drawing 2]****[Drawing 3]**



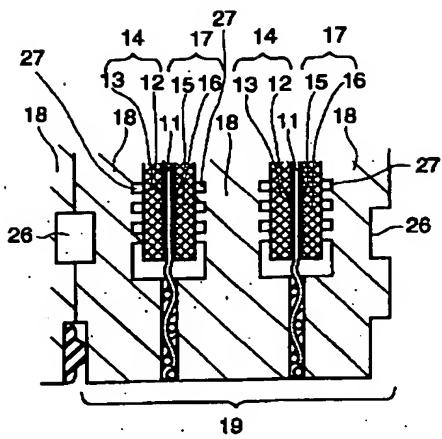
[Drawing 4]



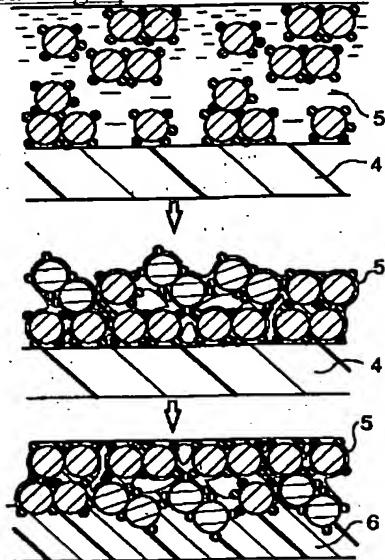
[Drawing 5]



[Drawing 6]



[Drawing 7]



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